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RESEARCH LABORATORY OF ELECTRONICS
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ABSTRACT

A new method using microwave cavity techniques has been developed for the study of the absorption of radiation in the radio frequency region by atoms and molecules. The present paper describes the application of this method to the study of the hyperfine structure of the ground state of Cs^{133}. Cesium vapor is introduced into a high Q resonant cavity which in turn is used to stabilize a microwave oscillator. The cavity is tuned to within a few megacycles of the hfs $\Delta\nu$. By application of a magnetic field the absorption line is split into Zeeman components, any one of which may be brought near to the resonant frequency of the cavity, causing a shift in this frequency due to the change in magnetic permeability accompanying the absorption. Incorporation of a small 30-cps component in the applied field causes a 30-cps frequency modulation of the oscillator stabilized to the cavity. This modulation may be detected by a suitable radio receiver. A study of the Zeeman components taking into account second order terms in the Breit-Rabi formula, enables us to evaluate $\Delta\nu$ as 9192.513 Mc/sec with a probable error no greater than 0.030 Mc/sec. A previous measurement by Millman and Kusch of 9192.6 $\neq$ 0.5 Mc/sec using molecular beam techniques is in excellent agreement with this value.

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MICROWAVE MAGNETIC ABSORPTION SPECTRUM OF ATOMIC CESIUM*

I. INTRODUCTION

Previous to the development of microwave techniques, information concerning interaction energies of nuclear magnetic moments with the extranuclear structures, nuclear spins, and g-factors could be obtained only by the methods of optical spectroscopy or atomic and molecular beams. The former is not capable of high precision, while the latter involves elaborate apparatus. At the time this research was begun (1945), microwave oscillators had not as yet been incorporated in such apparatus;[1] information concerning interaction energy was obtained by calculations based upon low-frequency data. As these are virtually extrapolations, the results, although surprisingly good, were not capable of the highest accuracy. Microwave spectroscopy, on the other hand, affords a relatively simple and precise method of obtaining this information.

The transitions produced by reorientation of nuclear magnetic moments in laboratory magnetic fields correspond to frequencies well below the microwave range. These may be studied by the method of nuclear induction[2] and magnetic resonance[3] experiments, which are now widely known. In the case of atoms whose total angular momentum is not zero ($J \neq 0$) there is produced at the nucleus by the electrons a magnetic field much larger than can be produced in the laboratory, and the frequency of radiation absorbed as the nucleus is reoriented in such a field is much higher, i.e. in the ultra-high-frequency or microwave range. In the presence of an external field, such absorption lines are split into a number of components by the Zeeman effect at weak fields or by the Back-Goudsmit effect at high fields. The nuclear spin may be determined by observation of the number of lines with zero field and with a field present, while the nuclear magnetic moment may be determined from the variation of frequency with magnetic field at moderate or strong fields.

In the case of atoms having an electronic ground state with $J = 1/2$, this ground state is split into two states of total angular momentum $F = I + 1/2$ and $F = I - 1/2$, where I is the nuclear spin. The energy difference between these is called the hyperfine-structure separation and will be denoted by $\Delta\nu$. In the presence of a weak magnetic field, each of these levels is

*A preliminary report of this work has been published by A. Roberts, Y. Beers, and A. G. Hill Phys. Rev. 70, 112 (1946).

[1] J. E. Nafe, E. B. Nelson and I. I. Rabi, Phys. Rev. 71, 914 (1947); D. E. Nagle, R. S. Julian and J. R. Zacharias, Phys. Rev. 72, 971 (1947).

[2] F. Bloch, W. W. Hansen and M. Packard, Phys. Rev. 70, 474 (1946); F. Bloch, Phys. Rev. 70, 460 (1946).

[3] E. M. Purcell, H. C. Torrey and R. V. Pound, Phys. Rev. 69, 37 (1946); N. Bloembergen, E. M. Purcell and H. C. Torrey 73, 679 (1948).

split into 2F + 1 components. Since the nuclear magnetic moment is small compared to that of the electrons, the spacings in the two sets of levels are very nearly equal and, to a good approximation, correspond to a g-factor of $1/(I + 1/2)$. Transitions between these levels take place according to the selection rules $\Delta m = \pm 1$ (π-polarization) and $\Delta m = 0$ (σ-polarization), where m is the magnetic quantum number associated with F and has 2F + 1 possible values ranging from +F to −F. There are transitions for all values of Δm ending on the 2I lower set of levels ($F = I - 1/2$). However, because of the virtual equality in spacing in the upper and lower sets of levels, all of the π-lines are doublets except for the extreme ones, $m = I - 1/2$ to $m = I + 1/2$ and $m = -(I - 1/2)$ to $m = -(I + 1/2)$. Thus, there are 2I + 1 π-lines and 2I σ-lines; and if the number of lines is counted, the nuclear spin I may be determined.

The subject of the present experiment was Cs^{133}. This had been investigated by optical spectroscopy[4] and by the Molecular Beam Magnetic Resonance Method[5] with the following results:[6] nuclear spin I = 7/2, magnetic moment = + 2.558 ± 0.007 nuclear magnetons, $\Delta\nu = 9192.6 \pm 0.5$ Mc/sec. Since the electronic structure has a ground state of $^2S_{\frac{1}{2}}$, it is split into levels having F = 4 and 3, respectively, yielding, in accordance with the previous remarks, a Zeeman pattern of 15 lines as indicated in Figures 1 and 2.

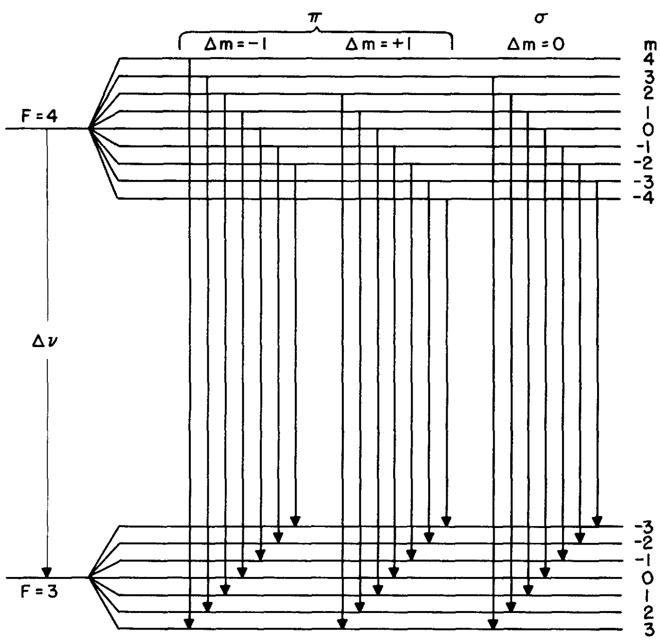

Fig. 1 Hyperfine-structure energy levels of Cs^{133} in a weak magnetic field.

[4] D. A. Jackson, Proc. Roy. Soc., A121, 432 (1928); L. P. Granath and R. K. Stranathan, Phys. Rev. 46, 317 (1934) and 48, 726 (1935); H. Kopferman, Zeits f. Phys. 73, 437 (1932).

[5] V. W. Cohen, Phys. Rev. 46, 713 (1934); S. Millman and J. R. Zacharias, Phys. Rev. 51, 1049 (1937); P. Kusch, S. Millman and I. I. Rabi, Phys. Rev. 55, 1176 (1939).

[6] J. B. M. Kellogg and S. Millman, Rev. Mod. Phys. 18, 323 (1946).

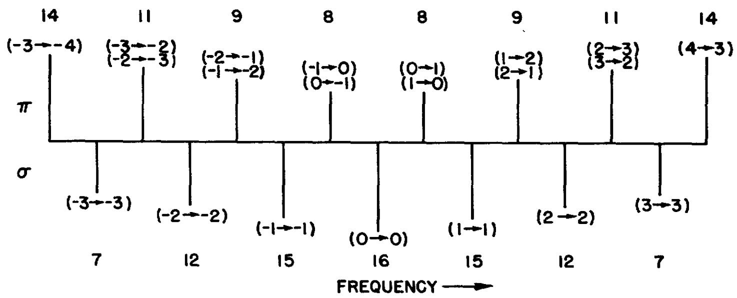

Fig. 2 Spectrum of Cs^{133} in weak magnetic field. Numbers in parentheses indicate quantum numbers of levels involved. Other numbers indicate relative intensity.

The relative intensities as calculated by the sum rules[7] are also indicated in Figure 2. It should be remembered that the π-lines may be excited in varying intensities relative to the σ lines (especially in the present experiment).

II. METHOD

One of the objectives of this experiment was to develop a method which could be applied to substances available in limited quantity, such as those produced by nuclear disintegrations. For such a purpose, a cavity system has an advantage over the more widely used waveguide systems and was a logical choice. In the present system, the frequency of a reflex velocity-modulation oscillator tube is stabilized (by means of a circuit due to Pound[8]) to the resonant frequency of a high-Q resonant cavity which contains the cesium vapor (see in Figure 3), the frequency being set a few megacycles

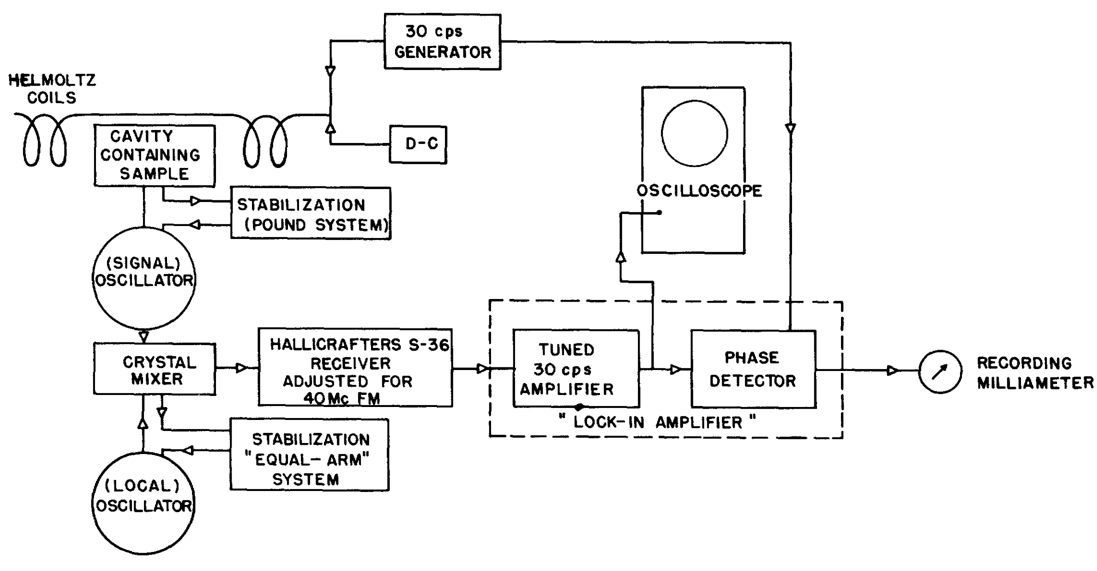

Fig. 3 Block diagram of apparatus.

[7] H. E. White, "Introduction to Atomic Spectra", McGraw-Hill Book Co., (New York, 1934) p.161.

[8] R. V. Pound, Rev. Sci. Inst. 17, 490 (1946) and 18, 132 (1947); Proc. I.R.E. 35, 1405 (1947); "Technique of Microwave Measurements", (edited by C. G. Montgomery) Radiation Laboratory Series, vol. 11, McGraw-Hill Book Co. (1947) (R. V. Pound and D. R. Hamilton) Ch. 2, p. 21.

away from the undeviated $(0 \to 0)$ line. The cavity is placed in the field of a pair of Helmholtz coils through which is flowing a d-c current and upon which is superimposed a small 30-cps a-c current whose amplitude is not greater than a few per cent of the d-c current. By means of the resulting steady magnetic field, one of the Zeeman absorption lines can be brought on or near to the frequency of the oscillator, resulting in (a) a reduction in the Q of the cavity due to absorption by the vapor and (b) a change in the resonant frequency of the cavity (except when the two resonant frequencies coincide exactly) because, near resonance, the vapor has a permeability differing from unity. The stabilization system is insensitive to the former effect but, due to the latter, there is a shift in the resonant frequency of the cavity. The effect of the a-c component of the magnetic field is to cause a periodic variation in the frequency (that is, a frequency modulation) of the oscillator at a rate determined by the frequency of the a-c field. The coincidence of the resonant frequencies of the absorption can be detected by feeding some of the output power of the oscillator into a suitable FM radio receiver and observing an a-f output signal of the frequency of the a-c magnetic field. An alternative method (hereafter called the "thermal drift method") was (with constant d-c field) to observe the output of the receiver as a slow drift of the resonant frequency of the cavity was produced by making a sudden small change in the current through electric heaters mounted on the cavity. The latter method was especially useful in determining line shapes. The line shape, that is, the dependence of receiver output on d-c field or frequency, will be shown later in this paper to be that of the derivative of a dispersion curve.

The FM radio receiver was a double-conversion superheterodyne. The signal was mixed in a silicon crystal with the output of another oscillator whose frequency was stabilized by an "equal-arm circuit"[9] to the resonant frequency of a standard wavemeter cavity tuned some 40 Mc/sec higher. A discussion of stabilization circuits will be given later. The output of the mixer went into a Hallicrafters S-36 superheterodyne receiver, which, tuned to 40 Mc/sec, served as the i-f amplifier and detector. An output was taken directly in parallel with the detector load resistance and went into a one-stage twin "T" narrow-band amplifier whose output went to either a standard oscilloscope with 60-cps sine or sawtoothed sweep, or a phase detector with a large time constant in the output. The latter effectively narrowed the overall bandwidth to a fraction of a cycle, yielding an improvement in signal-to-noise ratio. The tuned amplifier and phase detector were constructed as a single unit and, as such, constituted what is often called a "lock-in"

[9] W. G. Tuller, W. C. Galloway, and F. P. Zaffarano, Proc. I.R.E. 36, 794 (1948).

amplifier.[10] The output of this unit could actuate either a standard three-inch 0 – 1 milliameter or an Esterline-Angus recording milliameter. The recording milliameter, besides being essential in the thermal-drift method, was invaluable in the variable-field method in the earlier part of the work when the sensitivity was somewhat poorer than at the end. The output at some value of the field could be recorded for some minutes and then averaged by drawing a horizontal line through the trace to include subjectively as much extra area as was cut off in the peaks. A similar procedure would then be repeated at other values of the field. In this way, the detectability of weak signals could be greatly improved by averaging out fluctuations over such a great time.

Other cavity methods have been developed.[11,12] The present method yields a high sensitivity for several reasons: (a) reduced oscillator noise due to stabilization, as discussed in the following paragraph; (b) the narrow band-width and long observation time effected by the "lock-in" amplifier and recording milliameter, and (c) the modulation of the effect being investigated, making it easier to distinguish from spurious effects, and resulting in an improvement in what Goldman calls "generalized selectivity".[13] The analogous use of a modulated Stark effect has subsequently been widely used to improve sensitivity and generalized selectivity in waveguide systems.[14] On the other hand, the present method suffers the disadvantage of operating on one of the Zeeman components, resulting, in the case of cesium, in reducing by a factor of 15 the intensity which could be obtained from a given sample of vapor if observations were made on the absorption with no external field. In its present form, the present method can not detect the undeviated line either with or without modulation, although it could be detected by either of two modifications: (a) using a large a-c magnetic field and tuning the narrow a-f amplifier to twice the modulation frequency and using a reference voltage of this double frequency on the phase detector; (b) with zero magnetic field, introducing the modulation voltage on the mixer crystal according to a method originally suggested by Pound.[15] With the former, an improvement

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[10] The actual circuit is due to R. Beringer, M.I.T. Radiation Laboratory Drawing No. B-14292-A.

[11] B. Bleaney and R. P. Penrose, Proc. Roy. Soc. A189, 358 (1947).

[12] C. K. Jen, Phys. Rev. 73, 1248 (1948).

[13] S. Goldman, "Some Fundamental Considerations Concerning Noise Reduction and Range in Radar and Communication", Proc. I.R.E. 36, 584 (1948).

[14] R. H. Hughes and E. B. Wilson, Phys. Rev. 71, 562 (1947).

[15] R. V. Pound, Rev. Sci. Inst. 17, 490 (1946).

in sensitivity could undoubtedly be obtained, although it would be somewhat less than the factor of 15 since the audio output of the receiver would contain energy distributed in appreciable amounts in several Fourier components, the strongest being the second harmonic and the fundamental being entirely missing. In the present method, with small a-c field, practically the entire output is contained in the fundamental. The latter method suggested by Pound suffers by eliminating the improvement in generalized selectivity afforded by modulation of the effect being investigated.

The reduction in noise of the oscillator by stabilization requires further discussion. In the first place, it should be said that shot effect causes the oscillator tube to produce noise, as is well known. Near the frequency of oscillation, this is enhanced greatly by the presence of positive feedback. The large noise close to the principal frequency has been observed by Strandberg.[16] The noise can be thought of as a random fluctuation of the frequency and amplitude of the oscillations. This noise is produced by variations in the beam current and the number of electrons contained in each bunch. Equivalent fluctuations could have been produced by fluctuations of the tube voltages. However, it is widely known that reflex-velocity modulation tubes are very sensitive to frequency modulation and very insensitive to amplitude modulation by variation of tube voltages. Hence it can be inferred that the random frequency modulation produced by the noise is large and the amplitude modulation small. However, the stabilization circuit is designed to nearly eliminate fluctuations in frequency, especially those fluctuations of low frequency which are the most prevalent in the enhanced portion of the noise spectrum. This reduction in noise is observed experimentally.

III. APPARATUS

A. The Cavity

The cavity, as far as its r-f design is concerned, was copied after a standard wavemeter cavity which operates in a TE_{011} mode, except that the damping struts, which are normally included to suppress the TM_{111} mode, coincident with the TE_{011}, were omitted. The cavity was constructed of copper and brass and silver-plated.

The cavities used in the earlier part of the work were made vacuum-tight, and the cesium was distilled into them; they were then sealed off the pumps. Both waveguide and coaxial-line feeds with glass-to-metal seals were employed. The cavity was operated with its axis of symmetry vertical,

[16] M. W. P. Strandberg, Private Communication.

and a narrow glass tube was sealed to the center of the bottom to act as a cesium reservoir. A bifilar-wound electric heater on mica strips was fastened around the cavity and another electric heater wound on an asbestos tube surrounded the cesium reservoir. Copper-constantin thermocouples mounted on the bottom of the reservoir and the cavity were used for measuring the respective temperatures. The pressure was determined from a semi-empirical vapor pressure vs. temperature relation.[17] The cavity could be tuned by varying the position of the upper plate which was mounted on a threaded rod, operating through a flexible diaphragm of the type developed for tunable magnetrons.

These early models containing glass had very short lives before they sprang leaks, since hot cesium vapor readily attacks glass. In fact, only one successful run was made with cavities of this type, despite numerous attempts. The model which proved to be reasonably successful was not vacuum-tight but contained a replaceable quartz bottle which filled the useful volume of the cavity. A narrow quartz tube sealed to this bottle extended down through the bottom of the cavity to serve as a cesium reservoir in a manner similar to the glass reservoirs in the vacuum-tight cavities. Although these bottles did spring leaks, partly due to slow chemical action, their lifetime was satisfactory. This design had the minor advantage that the tuning mechanism, now being entirely in air, was much easier to operate and to adjust precisely. However, the design had several disadvantages. The Q was reduced to 10,000 or less, partly by the quartz and partly by oxidation of the metal surfaces, although the latter effect was reduced by maintaining a stream of nitrogen through the cavity. In addition, two strong modes and, occasionally, other weaker modes were observable. A more disturbing effect was that sometimes the high-Q mode had a discontinuity at one frequency, probably because of coincidence of its resonant frequency with that of another mode which was not otherwise observable. Such a discontinuity could be removed by rotating the quartz liner relative to the cavity. In addition, the introduction of the liner produced a mismatch, which was somewhat a function of the temperature. The latter difficulty was circumvented by the use at the cavity input of a slide-screw tuner which had its final adjustment at approximately operating temperature.

The cavity heater had bifilar winding of nichrome strip on a mica strip, which was wrapped around the cavity, and this was operated from a d-c source at about 2 amperes. Reversing the current produced no detectable change in frequency of the Zeeman lines, indicating that the average magnetic field of the heater was zero. However, it is possible that local inhomogeneities,

[17] R. W. Ditchburn and J. C. Gilmour, Rev. Mod. Phys. 13, 310 (1941).

causing broadening of the lines, may have existed, since the outside of the useful volume of the cavity was only 5 mm from the heater, while the separation of the bifilar nichrome strips was about one mm. This situation was more serious because the r-f field is strongest at the edge of the cavity. The largest source of experimental error on frequency measurements was due to drifts caused by thermal expansion of the cavity, since the wavelength is proportional to the linear dimensions of the cavity. A one-degree-centigrade change in temperature would change the frequency by 200 kc while, except for this effect, the electronic circuits are capable of maintaining the frequency constant within 50 or 100 cycles. It is quite possible that a much better heating system could be devised, employing the principle of thermal equilibrium with a boiling liquid such as napthalene or the melting of a solid such as tin, but this would have led to considerable complication of the apparatus.

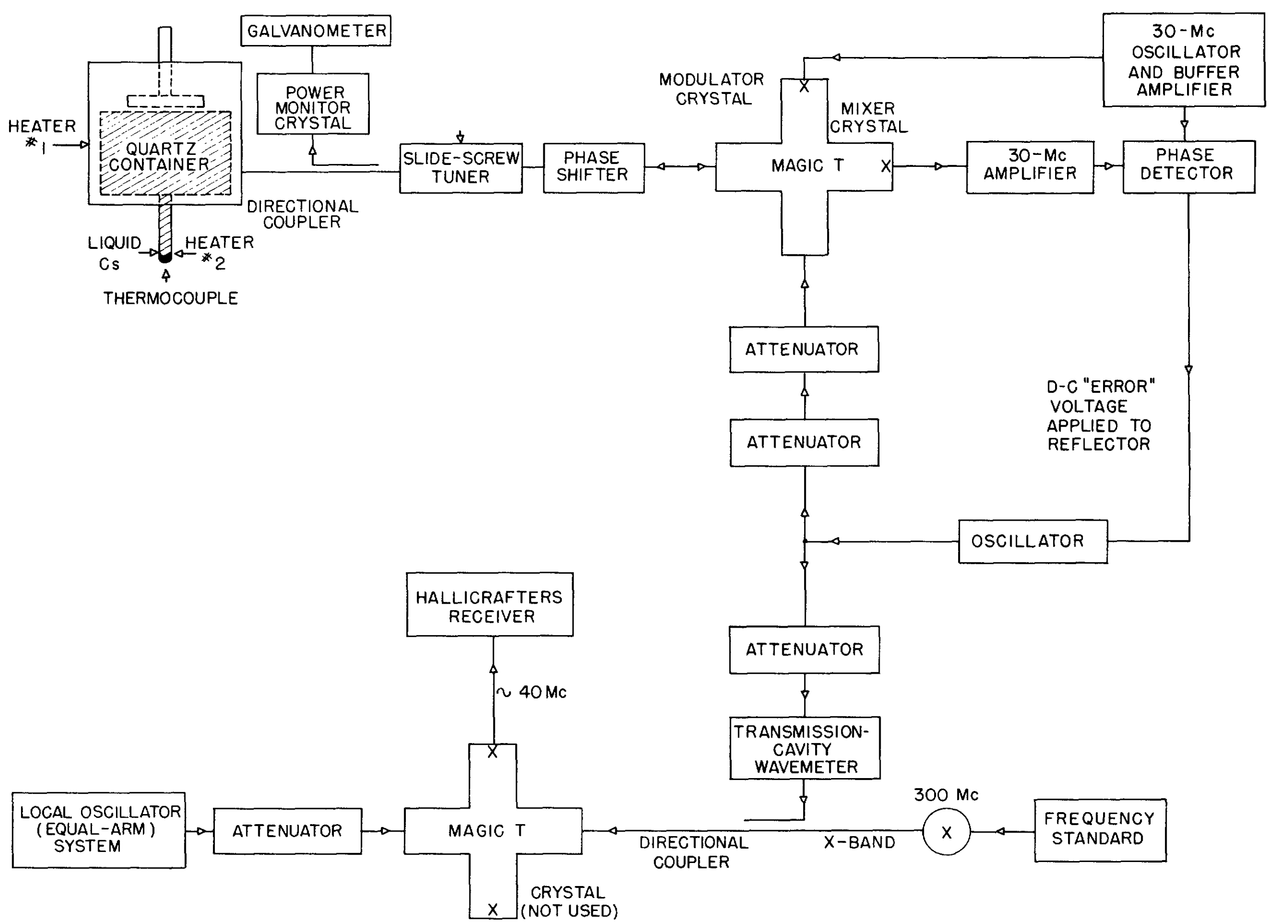

Fig. 4 Circuit details.

B. Stabilization Systems

It has already been mentioned that the signal oscillator employed the Pound system of stabilization[8] while the local oscillator employed the "equal arm" circuit.[9] The reason for the use of different circuits was that the power input to the cesium cavity had to be maintained at a low level to prevent saturation of the vapor, while no such restriction applies to the local oscillator circuit. It was discovered that the performance of the equal-arm circuit was inferior to the Pound circuit at low power levels because of noise, while at high levels the equal-arm circuit is to be preferred because of its ease in adjustment.

These circuits have been described adequately in the references which are cited herein, and the present remarks are confined to an explanation of why the Pound circuit is to be preferred at low levels and to a discussion of some of the adjustments.

In the Pound system, the error signal is derived by phase detection of a 30-Mc signal produced in a silicon crystal located at one arm of a magic T as indicated in Figure 5(a). This is proportional to the product of the amplitudes of a signal derived from the oscillator and a sideband signal produced by reflection of some of the oscillator signal around the magic T past the cavity and the modulator crystal. The circuit of the equal arm is identical, except that the position modulator and mixer crystals are interchanged. If E, ρ, k, and k' are the incident voltage, reflection coefficient

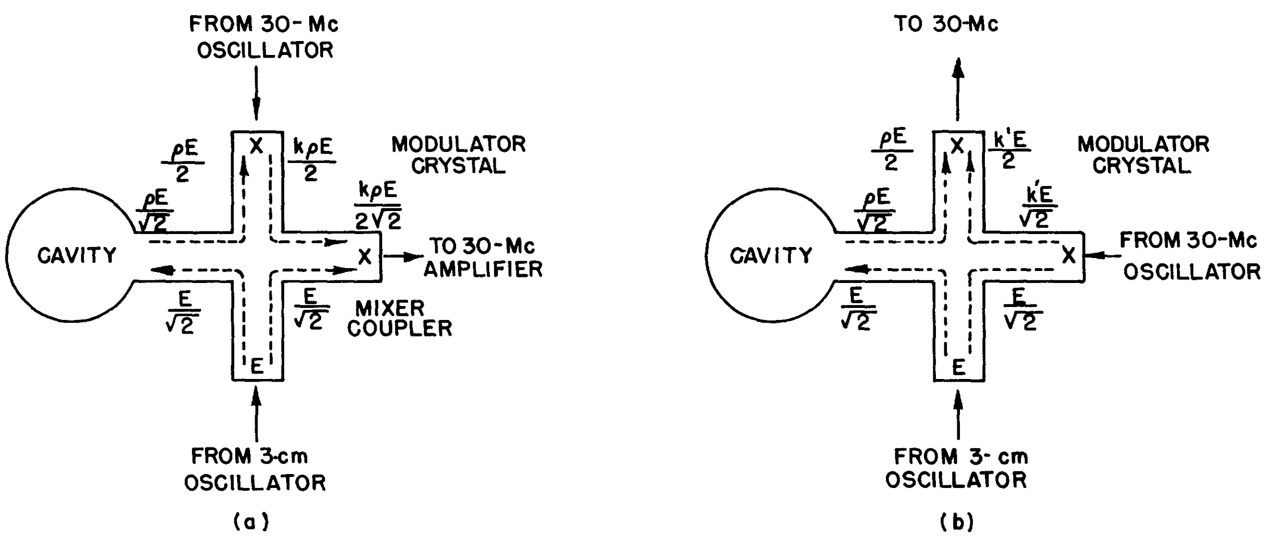

Fig. 5 Stabilization systems;
(a) Pound system, (b) equal-arm system.

of the cavity, and the modulation factors of the modulator crystal in the Pound and equal-arm systems, respectively, the voltages at various points have the values shown in Table I (as a consequence of the property of a magic T that the power incident at any arm is split between the two adjacent arms). In Figure 5, only the useful signals are indicated, and the perturbations by the multiple reflections of the unused signals are neglected. It is also

TABLE I

Voltages in Various Portions of Stabilization Systems

	Pound System	Equal-Arm System
Incident voltage	E	E
Voltage incident at cavity	$\frac{E}{\sqrt{2}}$	$\frac{E}{\sqrt{2}}$
Voltage incident at modulator crystal	$\rho\frac{E}{2}$	$\frac{E}{\sqrt{2}}$
Sideband voltage incident at mixer	$\frac{k\rho E}{2\sqrt{2}}$	$\frac{k'E}{2}$
Signal voltage incident at mixer	$\frac{E}{\sqrt{2}}$	$\rho\frac{E}{2}$
Product of sideband voltage and signal voltage at mixer	$\frac{k\rho E^2}{4}$	$\frac{k'\rho E^2}{4}$

assumed the impedances of the crystals are perfectly matched to their respective waveguides. The 30-Mc error signals are proportional to the quantities shown in the last line, which, for the same cavity power, would be equal if k were equal to k'. However, as ρ is small compared to unity, the modulator crystal in the Pound system operates at a lower incident power where it is more nonlinear and hence a more effective modulator; therefore k is greater than k'.[18] It can be seen, therefore, that when the power level is kept so low that noise in the i-f amplifier is not negligible compared to this 30-Mc error signal, the Pound system has one advantage of producing a greater error signal. In the second place, the modulator crystal is a source of noise which is cross-modulated on the sideband signals produced by it and transported by them to the mixer crystal. In the Pound system, since ρ is small compared to unity, the sideband signals are smaller than in the equal-arm system and less of this noise is transported to the mixer. In the ideal case, where the oscillator and cavity frequencies are exactly equal, ρ would be zero and none of this noise would appear at the mixer in the Pound system,

[18] Quantitative data on the modulation efficiency of silicon crystals is presented in the Quarterly Progress Report, Research Laboratory of Electronics, M.I.T., July 15, 1948.

while it would continue to appear there in the equal-arm system. At the same time, in this ideal case the signal power incident on the mixer crystal is larger than the sideband power in the equal-arm case, resulting in the 30-Mc impedance of the crystal remaining low and more nearly the value for which the input of the amplifier was designed.

The performance of the equal-arm system was so poor at low power levels that this experiment could not have been carried out had it been employed. Further improvement to the Pound system could have been effected by reinforcing the signal-frequency voltage at the mixer by feeding additional power via a supplementary route through directional couplers, a phase shifter, and attenuator connected between the oscillator and the mixer. Such a circuit was tried and found to have an improved performance but was critical in adjustment. It was not used in any actual experiments.

In both oscillator circuits, provision was included for sweeping the frequency of the tube by applying a small 60-cps signal to the reflector of the tube (when operated without stabilization) and observing the reflector voltage on an oscilloscope. In the equal-arm oscillator the various tuning adjustments, the phase shifter and screws on the crystal holders and the cavity of the oscillator tube, were varied to give a good discriminator curve with a crossover near the desired frequency; and when the reflector voltage switch was thrown to the stabilized position, the tube was found to lock-in with no trouble. This circuit was very reliable and often operated for two or three weeks without any adjustment other than setting the frequency to the desired value by tuning the wavemeter cavity. A range of about 20 Mc could be covered with no adjustment other than tuning this cavity.

No such simple procedure or reliable operation prevailed with the Pound circuit. It was not possible to tune by this oscilloscope method, since a reasonable locking pattern was not a sufficient criterion for "locking". Also the adjustments on the slide-screw tuner, the phase shifter, and modulator crystal mount hopelessly interlocked. First the crystal mounts were matched by removing them from their usual positions, connecting them through an attenuator to the oscillator which operated at the middle of the expected range of frequencies; these were tuned to give maximum crystal current. Then the crystal mounts were replaced, and their adjustments thereafter were not touched. Next the slide screw tuner was adjusted. The regular lead to the modulator crystal was disconnected and the oscilloscope was connected to it. A 60-cycle sweep was used on the oscillator and the scope pattern indicated the mode of the oscillator tube as reflected by the cesium cavity. The resonance of the cesium cavity was indicated by a notch in the scope pattern and its frequency could be measured by the coincidence of a much smaller notch caused by reaction of the transmission-type wavemeter.[19]

When the cesium cavity is matched to its guide, this notch extends down to the base line of the pattern, and the slide-screw tuner was adjusted to cause this notch to remain close to the base line as the phase shifter was varied throughout its range. (The phase shifter is the equivalent of adding an arbitrary length of line; if the cavity is perfectly matched, adding an arbitrary length of line will not change the match.) Finally the phase shifter was adjusted by the method described by Pound in his original papers, employing the voltmeter on the reflector voltage as an indicator.

C. Frequency Measurement

Frequency was measured with the microwave frequency standard constructed during the war by the M. I. T. Radiation Laboratory.[20] This has a probable uncertainty of 6 kc at 9000 Mc/sec, excluding experimental error of the comparison process. The final tripler to 900 Mc was not required. Output at approximately 300 Mc was fed to a silicon crystal connected to the apparatus as indicated in Figure 4. This crystal generated a harmonic at the signal frequency, an overall multiplication by a factor of 1488 from the fundamental of 6 Mc. Ambiguity from image response of the receiver and other harmonics was eliminated by checks using other multiplication factors and by comparison of the variation of fundamental frequency with variation in i-f frequency.

For comparison of the signal frequency with the standard, 60-cycle sweep was used on the local oscillator (normally "equal arm"-stabilized) and an oscilloscope was connected to the second detector of the Hallicrafters receiver operated on "narrow AM"; this combination became, in effect, an r-f spectrum analyzer. The tuning on the frequency standard was varied until the peak due to the frequency standard coincided with that due to the signal oscillator. Near coincidence the composite pattern has slow fluctuations, indicating low-frequency beats.

D. Magnetic Field

The working magnetic field was obtained from a round Helmholtz coil of conventional design, the diameter being about one foot, the resistance 1000

[19] This presentation was also very useful in checking the cavity and determining its modes. When the cavity had been recently filled, there was usually some liquid cesium in the cavity proper. This spoiled the r-f properties until it was distilled out by operating the cavity heater and leaving off the lower tube heater. This scope presentation was useful in monitoring this process.

[20] "Technique of Microwave Measurement" (edited by C. G. Montgomery), Radiation Laboratory Series, vol. 11, McGraw-Hill Book Co., (New York, 1947), Paragraphs 6.2 through 6.19.

ohms, and the impedance 2000 ohms at 30 cps. Since the diameter of the useful volume of the cavity was two inches, (which, compared to that of the coil, was not so small as could be desired), some broadening in the lines can be attributed to inhomogeneity in this field. The axis of this coil was placed parallel to either the vertical or the horizontal component of the earth's field, while the axis of the cavity was vertical, and the remaining component of the earth's field was cancelled with the aid of a square Helmholtz coil, one foot on a side (after an unpublished design of R. B. Lawrance). Correction for the effect of the component parallel to the main field was determined by observing the field necessary to bring some line on to the signal frequency and then reversing the field and finding the corresponding value. With the main field vertical, the σ-lines were favored over the π-lines by roughly 12 db over the relative values indicated in Figure 2 while, with the main field horizontal, the π-lines were favored by a similar amount.

The 30-cps modulation voltage was derived from a miniature motor generator consisting of a 1800-rpm 115-volt, 60-cycle, 7-watt synchronous motor (Bodine Electric Co., Chicago, Type KYC-22) and a two-pole permanent-magnet angle-phase a-c generator, 1800 rpm, 37 volts ("General Elnico", Electric Indicator Co., Stamford, Conn.). The output was amplified by two 6V6 tubes operating in a conventional push-pull circuit. The d-c and a-c voltages were fed to the Helmholtz coil by a conventional "parallel feed" network consisting of an inductance and a capacity. By having a moderately high impedance coil, this network could employ components of reasonable size.

By generating the 30 cps in this manner, coherently with the line frequency, unpleasant beats between the second harmonic and hum in the lock-in amplifier were avoided. The signal-to-noise ratio could be improved by the use of a higher modulation frequency as crystals in the circuit cross-modulate with the signal noise in an amount which increases with decreasing frequency. On the other hand, as the modulation frequency is increased, eddy-current losses increase and the a-c for a given power input decreases. The eddy currents in the cavity itself shield the interior; this shielding is nonuniform, so that the a-c field is not homogeneous within the cavity. Experiments indicated that, with the present apparatus, it would not be practical to raise the modulation frequency by more than a factor of 5.

IV. RESOLUTION

At weak field strengths, where the splitting is accurately proportional to the field strength, the following relation must hold if the center of the s'th line from the center of the Zeeman pattern is to coincide with the frequency of the stabilized oscillator:

$$\nu = \Delta\nu + s\alpha\, B_s \quad , \tag{1}$$

where

B_s = flux density
α = a constant (for Cs^{133} equal to 0.35 Mc/gauss, or very nearly one-quarter of a Lorentz unit).

If b is half-power line width, the values of the flux density B_{1s} and B_{2s} necessary to make the half-power points of the s'th line coincide with the frequency of the oscillator are given by

$$\nu = \Delta\nu + \tfrac{1}{2}b + s\alpha\, B_{1s} \tag{2}$$

and

$$\nu = \Delta\nu - \tfrac{1}{2}b + s\alpha\, B_{2s} \quad . \tag{3}$$

By algebraic combination of these equations it can be shown that

$$\begin{aligned} b &= (\nu - \Delta\nu)\left(\frac{B_{2s} - B_{1s}}{B_s}\right) \\ &= \delta\nu\left(\frac{B_{2s} - B_{1s}}{B_s}\right) \quad , \end{aligned} \tag{4}$$

where $\delta\nu$ = difference in frequency of the oscillator and central line.

Equation (4) yields the method for calculating line width from data taken by the fixed-frequency variable-field method. Since $\delta\nu$ is the same for all lines, Eq.(4) implies that, for all lines, the same fractional change in d-c magnetic field is required to cause the line to pass the frequency of the oscillator. It also implies that, for a uniform presentation of the lines, the a-c field should be kept a constant percentage of the d-c field.

The s'th line will be barely resolved when B_{1s} is greater than $B_{2(s+1)}$ and when $B_{1(s-1)}$ is greater than B_{2s}. By application of Eqs.(2) and (3), it can be shown that for resolution

$$|\delta\nu| > (|s| + \tfrac{1}{2})\, b \quad . \tag{5}$$

In the present work, all important measurements were made with lines corresponding to s = ± 1, or ± 2. The line width was 0.15 Mc/sec, and since $\delta\nu$ was always greater than 2 Mc/sec, resolution was always adequate.

V. LINE WIDTH

Energy is absorbed or radiated by the cesium atoms by the following processes: (a) spontaneous emission, (b) stimulated emission, (c) absorption,

(d) collision with other cesium or foreign gas molecules, and (e) collision with walls of the cavity. At microwave frequencies, spontaneous emission is negligible (as compared to optical frequencies) as the probability of emission (cf. Einstein's "A" coefficient in radiation theory) is proportional to the cube of the frequency. The net absorption of radiation at thermal equilibrium is the difference in the absorbed radiation and the radiation produced by induced emission. This is proportional to the total number of atoms and the relative population in the states $F = 3$ and $F = 4$. The relative populations of the two states, as given by the Boltzmann factor, is only about 0.1 per cent. Thus the difficulty of having a limited power dissipation by the finite number of molecules within the cavity is aggravated by the fact that only one atom in a thousand is effective in absorbing radiation. As collisions between two molecules and between a molecule and the walls are much more probable than spontaneous emission, they are mainly responsible for maintenance of the relative population of the atoms in the two states in equilibrium with the temperature.

In the present experiments, the temperature was approximately 500°K. The kinetic theory of gases gives for cesium atoms at this temperature an average velocity of 2.5×10^4 cm/sec. Since the smallest dimension of the cavity is 2 cm, in the absence of collisions with other atoms collisions with the walls would occur on the average in a time $t_w = 10^{-4}$ sec.

The effective value of the diameter of the cesium atom which determines how close the atoms must approach one another to disturb the radiation process is not necessarily the value which determines the macroscopic properties, such as the coefficient of viscosity and diffusion constant, since the latter requires the transfer of translational kinetic energy, while the former does not. However, if one assumes a value of the atomic diameter of 3×10^{-8} cm, which accounts for macroscopic properties of typical atoms and also assumes a pressure of 3×10^{-2} mm Hg and a temperature of 500°K (values which are typical for the experiment), the calculated mean time between collisions between cesium atoms is $t_c = 3.7 \times 10^{-5}$ sec.

Thus collisions between atoms, under these assumptions, are about three times as likely. Since the line width is the reciprocal of π times the collision time,[21] the corresponding line width would be about 10 kc.

Another natural cause of line broadening is the Doppler effect. From the average velocity of 2.5×10^4 cm/sec, one can compute that the average shift is about 8 kc and that the width due to this cause is about 16 kc.

The typical measured width under good conditions on singlet lines was about 150 kc. (The method of measurement will be described later.) The

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[21] J. H. Van Vleck and V. F. Weisskopf, Rev. Mod. Phys. 17, 227 (1945).

discrepancy of these calculated values with the measured is to be attributed to instrumental effects: inhomogeneity of the magnetic field, collisions with other molecules present as impurities, and power saturation effects. Since measurements were made with values of $\delta\nu$ in the vicinity of 5 Mc/sec, a 0.1 per cent inhomogeneity in the field could produce a broadening of 50 kc, and it is possible that the inhomogeneities were somewhat larger. The power dissipated in the vapor was near to saturation, although somewhat below, and probably some broadening is due to this cause.

VI. LINE SHAPE (THEORETICAL)

Van Vleck and Weisskopf[21] have derived a theoretical expression for the absorption coefficient of a gas whose line width is mainly due to collisions:

$$\gamma = \left(\frac{4\pi^3 \nu N}{3ckT}\right) \frac{\sum_{ij} \left[|\mu_{ij}|^2 \; \nu_{ij} f_{ij}(\nu_{ij}, \nu) \right] e^{-E_j/kT}}{\sum_j e^{-E_j/kT}} , \qquad (6)$$

where

μ_{ij} = matrix element connecting two states i and j,

ν_{ij} = frequency of transition connecting states i and j of energies E_i and E_j respectively,

ν = frequency of incident radiation,

N = number of molecules per cc,

$f(\nu_{ij},\nu)$ = "structure" factor governing the shape of the line, the expression for which is given later.

This equation contains a simplifying assumption, valid in the present case, that $\nu_{ij} << (kT/h)$, which makes it unnecessary to distinguish between

$$e^{-E_i/kT} \quad \text{and} \quad e^{-E_j/kT}$$

except where they are involved in a difference.

In the present case, if the external field is zero, there are only two states F = 3 and F = 4, and the summations contain only two terms. Since there is only one matrix element involved, it will be denoted by μ. The ν_{ij} has only one value: what has been called $\Delta\nu$ in our present notation. Then the absorption coefficient is given by:

$$\gamma = \frac{4\pi^3 \nu N |\mu|^2 \; \Delta\nu \; f_{ij}}{3ckT} . \qquad (7)$$

The expression for f_{ij} is:

$$f_{ij} = \frac{\upsilon}{\pi\Delta\upsilon}\left[\frac{\frac{b}{2}}{(\Delta\upsilon - \upsilon)^2 + (\frac{b}{2})^2} + \frac{\frac{b}{2}}{(\Delta\upsilon + \upsilon)^2 + (\frac{b}{2})}\right] , \qquad (8)$$

where b = line width (width at half maximum power).

In the present case, b (about 0.15 Mc/sec) is small compared to υ and $\Delta\upsilon$ (9192 Mc/sec). Under these conditions, the second term in Eq.(8) is negligible. Also (except where the difference $\Delta\upsilon - \upsilon$ is involved), υ can, with a good approximation, be considered a constant and equal to $\Delta\upsilon$. With these approximations,

$$\begin{aligned}\gamma &= \frac{4\pi^2 N|\mu|^2}{3ckT} \; \frac{2\upsilon^2 b}{4(\Delta\upsilon - \upsilon)^2 + b^2} \\ &= \frac{8\pi^2 N|\mu|^2 \upsilon^2}{3ckTb} \; \frac{1}{y^2 + 1} ,\end{aligned} \qquad (9)$$

where

$$y = \frac{2(\upsilon - \Delta\upsilon)}{b} . \qquad (10)$$

According to classical dispersion theory,[22] the dielectric constant is given by

$$\epsilon = 1 + \frac{\beta}{\Delta\upsilon^2 - \upsilon^2 - j\upsilon b} , \qquad (11)$$

where β is a constant which, for the present, will not be evaluated, and j is the square root of -1.

If the previous approximation and Eq.(10) are employed,

$$\epsilon = 1 + \frac{j\beta}{\upsilon b(1 + jy)} . \qquad (12)$$

This may be resolved into real and imaginary parts;

$$\epsilon = \epsilon' - j\epsilon'' ,$$

where

$$\epsilon' = 1 - \frac{\beta y}{b\Delta\upsilon(1 + y^2)} \qquad (13)$$

and

$$\epsilon'' = \frac{\beta}{b\Delta\upsilon(1 + y^2)} . \qquad (14)$$

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[22] See for example, J. C. Slater and N. H. Frank, "Electromagnetism", McGraw-Hill Book Co. (New York, 1947), pp. 107-108.

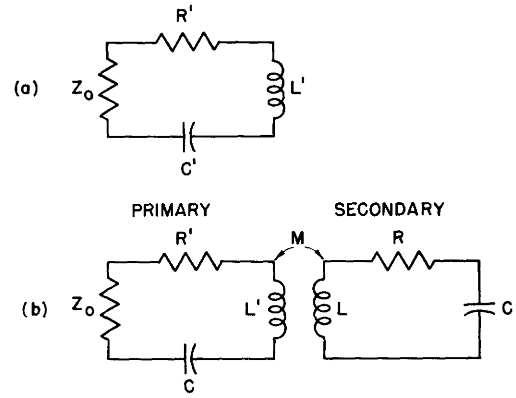

Fig. 6 Equivalent circuits of cavity: (a) without gas; (b) with gas.

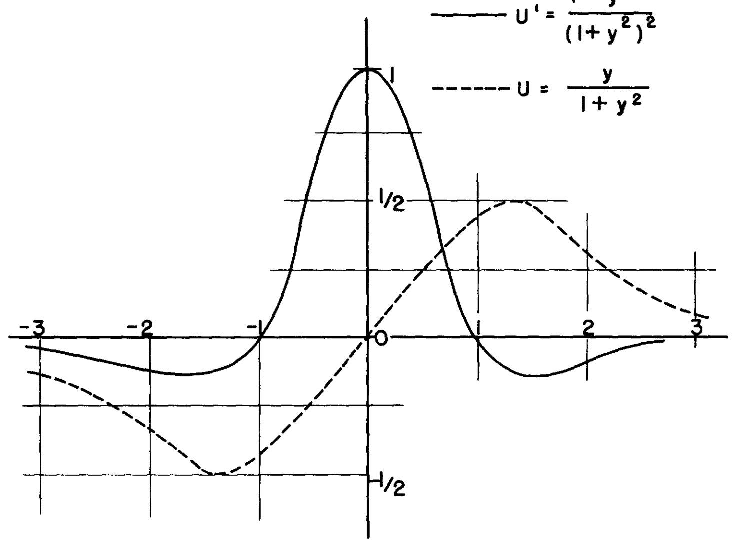

Fig. 7 Theoretical line shapes.

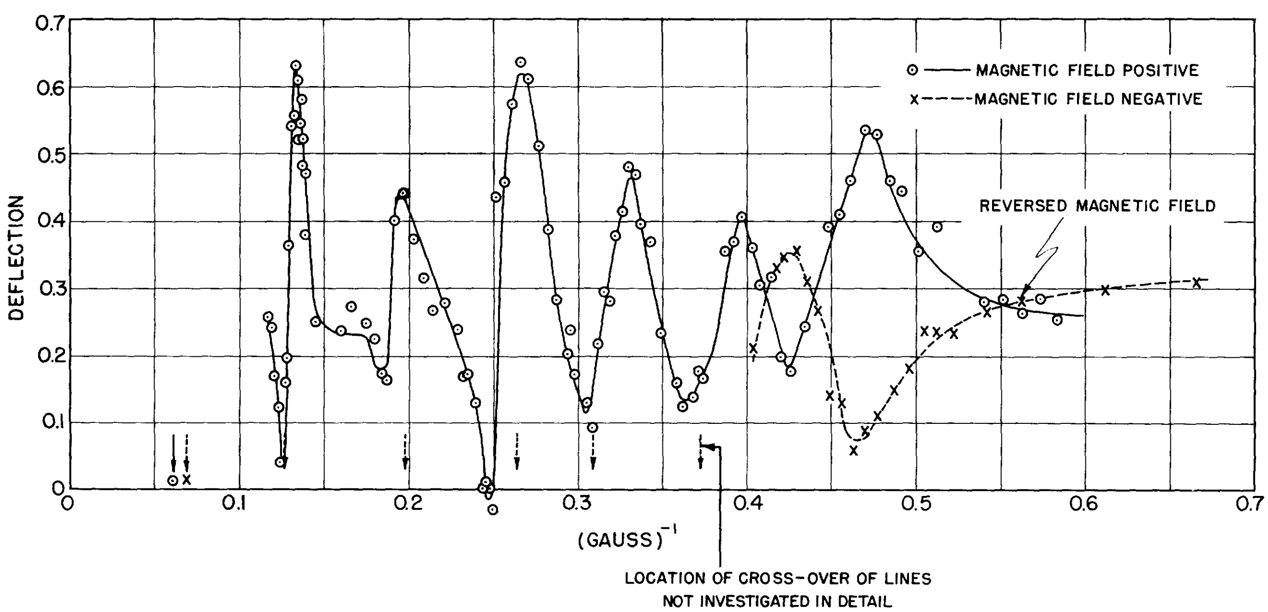

Fig. 8 Absorption of X-band radiation by cesium vapor: pressure, 4×10^{-2} mm Hg; frequency, 9197 Mc/sec.

The complex dielectric constant is equal to the square of the complex index of refraction. When ϵ'' is small compared to unity, as is the case for gases, the absorption coefficient is $(2\pi\nu/c)$ times the imaginary component of the index of refraction, or

$$\gamma = \frac{\pi\beta}{2cb(1 + y^2)} \quad . \tag{15}$$

The classical dispersion theory has been derived for electric dipole radiation, while in the present work we are concerned with magnetic dipole radiation. It is for this reason that the value of β from classical theory has not been given. It is to be assumed that the index of refraction would behave as a function of the frequency (or of y) in the same manner for the two types of radiation, and, therefore, it is not surprising that Eqs.(9) and (15) should show the same dependence upon y. We may therefore proceed to apply the classical dispersion theory to the present situation, provided that: (a) β is evaluated by equating the coefficients in Eqs.(9) and (15), and (b) we interpret ϵ as representing the magnetic permeability. By (a),

$$\beta = \frac{16\,\pi N|\mu|^2\nu^2}{3kT} \quad . \tag{16}$$

When an external magnetic field is applied, it is to be expected that the behavior of the gas near the absorption frequency of one of the Zeeman components, if completely resolved, would be correctly described by these theories, provided the following changes were made: (a) $\Delta\nu$ would be replaced by ν_0, the resonant frequency of the particular line (which, of course, is a function of the flux density), and (b) the value of β given by Eq.(16) is multiplied by a factor P, equal to the intensity of the particular line divided by the sum of intensities of all the components.

The stabilization circuit causes the oscillator tube's frequency to be that at which the imaginary part of the reflection coefficient of the waveguide feeding the cavity is zero. This frequency is modified by the presence of a material of magnetic permeability ϵ.

If a certain reference plane is chosen in this waveguide, the behavior of the cavity can be shown,[23] near a resonant frequency, to be equivalent to the lumped constant circuit shown in Figure 6(a). In this, Z_0 is the characteristic impedance of the guide, while L', C', R' are, respectively, the inductance, capacitance, and resistance associated with the cavity (without the effect of the gas). The total impedance in this circuit is:

[25] See for example, "Principles of Microwave Circuits", (edited by C. G. Montgomery) Radiation Laboratory Series, vol. 8, McGraw-Hill Book Co. (New York, 1947), pp. 228-230.

$$Z' = Z_o + R' + j(2\pi\nu L' - \frac{j}{2\pi\nu C'}) \quad . \tag{17}$$

According to a standard treatment, this may be written

$$Z' = (R' + Z_o)(1 + jy') \quad , \tag{18}$$

where

$$y' = Q'(\frac{\nu}{\nu_o'} - \frac{\nu_o'}{\nu}) \quad . \tag{19}$$

The resonant frequency is

$$\nu_o' = \frac{1}{2\pi\sqrt{L'C'}} \quad . \tag{20}$$

The loaded Q is

$$Q' = \frac{2\pi L'\nu_o'}{R' + Z_o} \quad . \tag{21}$$

Also it can be shown that the half-power bandwidth is

$$b' = \frac{\nu_o'}{Q'} \quad . \tag{22}$$

In the case that $Q' \gg 1$, or $\nu_o' \gg b'$, Eq.(19) becomes approximately equal to

$$y' = \frac{2(\nu - \nu_o')}{b'} \quad . \tag{23}$$

The effect of the gas upon the cavity can be represented by multiplying L' by the value of ϵ from Eq.(12). When this has been done, it can be shown that the equivalent impedance becomes:

$$Z'' = Z' + \frac{2\pi\beta L'}{b(1 + jy)} \quad . \tag{24}$$

The similarity in form of y in Eq.(10) to y' in Eq.(23) is, of course, obvious. Therefore, the denominator of the second term on the right of Eq.(24) can be seen by reference to Eq.(8) to have the same dependence upon frequency as a series-tuned L-R-C circuit. In fact (especially if one remembers that in Eq.(16) β contains the square of the frequency as a factor), Z" has the same dependence upon frequency as the equivalent impedance of the primary of circuit shown in Figure 6, which is known from standard circuit theory to be

$$Z'' = (Z_o + R')(1 + jy') + \frac{4\pi^2\nu^2M^2}{R(1 + jy)} \quad . \tag{25}$$

Thus the cavity, with the effect of the gas included, can be represented by the equivalent circuit shown in Figure 6(b).

The imaginary part of the reflection coefficient is zero at the frequency at which the reactive component of Z" is zero. Placing the reactive component of Z" equal to zero yields the following condition:

$$\upsilon - \upsilon_o' = - \frac{2\beta}{b} \frac{y}{1 + y^2} \quad . \tag{26}$$

Of course, y is proportional to υ (see Eq.(10)), and thus this equation gives υ as a cubic function of υ_o, which replaces $\Delta\upsilon$ as the resonant frequency of the gas, and of υ_o' (the resonant frequency of the cavity). The general solution of this equation is difficult. However, as will be shown later, $\upsilon - \upsilon_o'$, the shift in oscillator frequency due to the gas, is very small. Therefore, in the expression for y, we may approximate υ by υ_o':

$$y = \frac{2(\upsilon_o' - \upsilon_o)}{b} \quad . \tag{27}$$

Since, for weak field, υ_o' is proportional to the flux density B, the dependence of the frequency shift of the oscillator upon magnetic field is proportional to the function

$$U = \frac{y}{1 + y^2} \quad . \tag{28}$$

This function is plotted in Figure 7. It has extreme values of $\pm$ 1/2 at $y = \pm 1$, at which the absorbed power is one-half maximum.

However, the 30-cps output of the FM receiver is proportional to the rate of change in frequency with magnetic field; therefore the dependence of receiver output with d-c magnetic field is proportional to

$$U' = \frac{dU}{dy} = \frac{1 - y^2}{(1 + y^2)^2} \quad . \tag{29}$$

This function also is plotted in Figure 7. It has a maximum of 1 at $y = 0$ (line center), zeroes at $y = \pm 1$ (half-power points of the absorption curve), and minima of -1/8 at $y = \pm \sqrt{3}$.

VII. FREQUENCY SHIFT OF OSCILLATOR

As a final step, it is desirable to calculate the maximum possible frequency shift and to relate it to the absorption coefficient, a quantity which is directly measured by other methods of microwave spectroscopy and, therefore, is of great interest.

From Eq.(26), the maximum frequency shift can be seen to be (by placing $y = |1|$)

$$|\upsilon - \upsilon_o'|_{max} = \frac{\beta}{b} \quad . \tag{30}$$

The maximum value of the absorption coefficient is found by placing $y = 0$ in Eq.(15), yielding:

$$\gamma_{max} = \frac{\pi\beta}{2cb} \quad . \tag{31}$$

By eliminating β/b between these two equations,

$$|\nu - \nu_o|_{max} = \frac{2c}{\pi} \gamma_{max} \quad . \tag{32}$$

The γ_{max} is calculated numerically by placing $y = 0$ in Eq.(9) and multiplying the value there by a factor P to account for the reduction in intensity caused by splitting of the original level into its Zeeman components (i.e., P is the ratio of the intensity of the particular component to the sum of the intensities of all components). In the following calculation, an average value of 1/15 will be used.

The value of $|\mu|^2$ can be shown to be $(63/4)\ \mu_o^2$, where μ_o is the Bohr magneton.[24]

If one substitutes the additional following numerical values into Eq.(9),

$N = 5.4 \times 10^{13}$ atoms/cc, corresponding to a pressure of 3×10^{-2} mm and a temperature of 500°K,
$b = 1.5 \times 10^5$ cps,
$\nu = 9.2 \times 10^9$ cps,
$T = 500°K$,

as well as standard values of c, k, and μ_o, he obtains a value

$$\gamma_{max} = 3.5 \times 10^{-8}/\text{cm} \quad .$$

The corresponding value of the frequency shift is

$$\nu - \nu_{o\ max} = 6.5 \times 10^2 \text{ cps} \quad .$$

Experimentally, the frequency shift is somewhat smaller. In fact, if

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[24] See footnote No. 6 in the paper by J. H. Van Vleck, Phys. Rev. 71, 413 (1947); also E. U. Condon and G. H. Shortley, "Theory of Atomic Spectra", Cambridge University Press (New York, 1935) pp. 64–72.

Quantities in Condon and Shortley's notation have the following values:

$$j_1 = I = 7/2 \quad ,$$
$$j_2 = J = 1/2 \quad ,$$
$$j = F = 4 \quad ,$$
$$j' = F = 3 \quad .$$

$(\gamma j_1 j_2 j \vdots J_1 \vdots \gamma j_1 j_2 j')$ is evaluated by Eq. (2b) p. 66.

$\langle\vdash\!\dashv\rangle\ (j,j-1)$ is evaluated by Eq.(2) p. 71.

one listens to the audio-frequency beat produced by running the Hallicrafters receiver on AM with the beat oscillator tuned on, one cannot detect an audible change in pitch as the magnetic field is varied. The explanation for the discrepancy is not clear. It is known that the stabilization circuit causes the tube's frequency to be essentially that of the cavity, for when a calibrated wavemeter is used as the stabilization cavity, the output frequency agrees to at least one Mc of the indicated frequency; and if the resonant frequency of the cavity is varied by five or ten Mc, there is a corresponding change. The shift must, therefore, be at least a factor of 10 smaller. At any rate, the frequency shift is small enough to justify the previous approximate method of calculating line shape. Therefore, if the observed shift in frequency of the tube is smaller than the theoretical value, it is because the change in resonant frequency of the cavity is smaller than the theoretical value. The only experimental factor which can reasonably have some bearing on this discrepancy is the possibility of power saturation of the gas, which would effectively reduce the absorption coefficient.

VIII. POWER SATURATION

Most measurements were made with a mixer-crystal current of 1 to 3 microamperes. This would indicate a power incident upon the cavity of about 10^{-6} watt. Saturation was then at about 10^{-5} or 10^{-6} watt.

Of course, not all of the power is dissipated within the vapor. The fraction dissipated within the vapor can be determined by use of the theory of the equivalent circuit; in fact, it is given by the real (resistive) part of the coupled impedance, the second term on the right of Eq.(24), to the total resistance of the cavity, which is essentially R'. By expressing L' in terms of $(R' + Z_o)$ and Q' through the use of Eq.(21), assuming $R' = Z_o$ and by expressing β/b in terms of γ_{max} by use of Eq.(32), it can be seen that this fraction is

$$\frac{2cQ'}{\pi\nu}\,\gamma_{max}$$

or about 3.5×10^{-4} if one uses a value of Q' of 5000 and other values as before. Thus, about 10^{-9} watt is dissipated within the vapor.

This value yields information concerning the relaxation time t_r in the gas. The saturation power must be equal to the total energy which can be stored in the gas divided by the relaxation time. The total energy which can be stored is the product of the following quantities: (a) the number of atoms per cc (5.4×10^{13}); (b) the volume of the cavity (50 cc); (c) the Boltzmann factor (10^{-3}), and (d) the energy per quantum $(6 \times 10^{-24}$ joules) or 1.6×10^{-11} joules. Therefore the relaxation time is about 10^{-2} sec, while the average time between "collisions" is of the order 10^{-4} sec. Thus an atom makes on the order of 100 collisions before a transition occurs.

IX. EXPERIMENTAL LINE SHAPE

The first measurements made on April 30, 1946 with the constant-frequency variable-field method with a glass window cavity yielded the results shown in Figure 8. To have a presentation resembling that similar to optical spectroscopy, the output of the lock-in amplifier is plotted against reciprocal of the magnetic field, the a-c field being a constant (rather than a constant percentage as indicated in Section IV). In this figure, six lines on the high-frequency side of the undeviated line are shown in detail, and the position of the seventh (which could not be investigated in detail because of overload of the Helmholtz coils) is indicated.

It is to be noted that, as expected, the lines occur at equal intervals but do not have the expected shape, that is, they do not resemble a plot of the function U' but rather resemble the function U. If the same data are plotted on a semi-logarithmic scale, it can be shown that the lines have apparent equal widths, as predicted in the theory of resolution (see Sec. IV).

No further measurements were made until the summer of 1948, when quartz-lined cavities were used. Many measurements were made, using both the constant-frequency and thermal-rate-of-drift methods. The line shapes always agreed, more or less, with the theoretical shape, that is, with a plot of the function U', although generally one or the other negative peak was larger than could be expected and the other smaller (the discrepancy undoubtedly being due to adjustments of the stabilizer). An observation of line shape is shown in Figure 9, in which the negative peaks happened to be nearly equal. The ratio of positive-peak height to negative-peak height was 5.9, as compared with a theoretical value of 8. The ratio of the separation of the negative peaks to the separation of the zeroes is 1.6, as compared to a theoretical value of 1.7. Cutting in or out the transmission type wavemeter or putting a large amount of attenuation at the input of the Hallicrafters receiver causes no appreciable change in the shape of the line. Thus there is good agreement between theoretical and experimental line shapes.

It was difficult to account for the anomalous line shape observed in 1946 (and shown in Figure 8) until it was remembered that at that time the gain control on the stabilizing circuit was set for the lowest possible gain at which the circuit would operate. When the gain control was set in this manner, line shapes similar to that in Figure 8 were observed as indicated by the solid curve in Figure 10. With somewhat lower gain, the resemblance was more complete, although the tube jumped out of stabilization

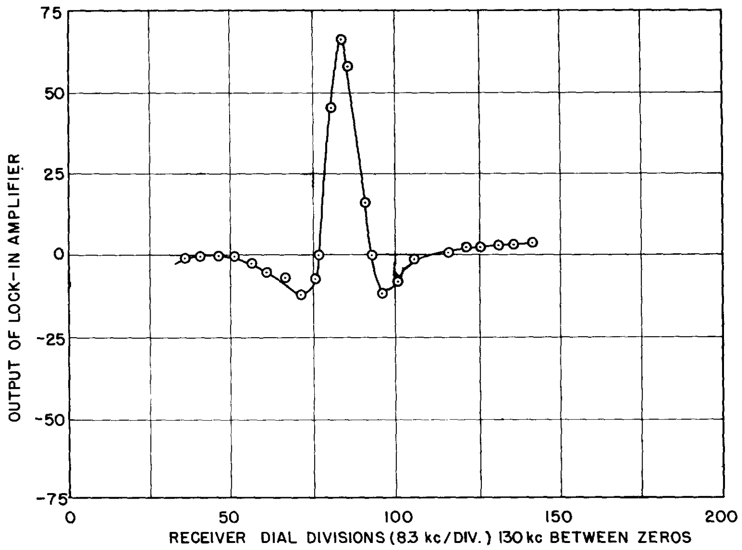

Fig. 9 Thermal-drift method ($-1 \to -1$) line (π): pressure, 2.8×10^{-2} mm Hg; d-c field, 8.2 gauss; a-c field 5×10^{-2} gauss rms; no wavemeter.

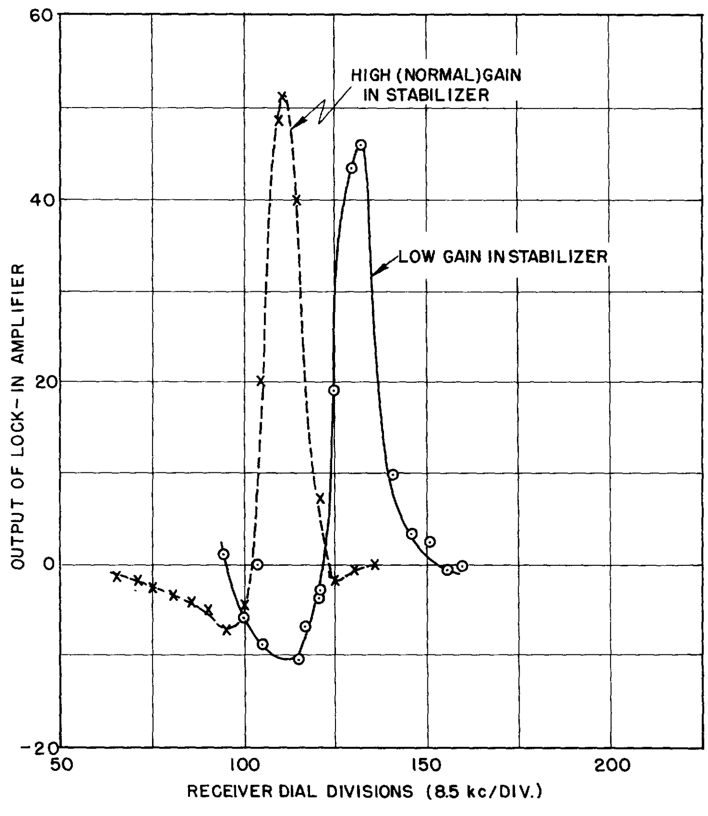

Fig. 10 Thermal-drift method ($0 \to -1$, $-1 \to 0$) line (σ): pressure 2.9×10^{-2} mm Hg; d-c field, 12.6 gauss; a-c field, 0.1 gauss rms.

before complete data could be taken. With high gain, the second negative peak appeared as indicated in the dashed curve in Figure 10. (Here, however, is an extreme case of unequality of negative peaks.) This experiment was repeated by the constant-frequency variable-field method with the same result.

It is to be noted that the zeroes of a U'-type curve correspond to the half-power points of the absorption curve. Thus the line width can be determined by recording the difference in frequency at the two zeroes, or, in the constant-frequency method, the values of the flux density corresponding to the zeroes and substituting these into Eq.(4); these yield values of 130 to 200 kc.

In all the 1948 experiments, measurements were made with both phases of reference voltage on the lock-in amplifier and the results shown are the differences in these values. In this way, zero drift and nonlinearities in the lock-in amplifier were largely eliminated.

X. MEASUREMENT OF $\Delta\nu$

The energy of any level involved in the present experiment is given the Breit-Rabi formula[25], applied to an atom in a $^2S_{\frac{1}{2}}$ state:

$$E_{i \pm \frac{1}{2}, m} = -\frac{h\Delta\nu}{2(2I+1)} + g_1\mu_o Bm \pm \frac{h\Delta\nu}{2}\left(1 + \frac{4m}{2I+1}x + x^2\right)^{1/2}, \qquad (33)$$

where

$E_{i \pm \frac{1}{2}, m}$ = energy of a level having total angular momentum $F = I \pm 1/2$,

m = magnetic quantum number,

μ_o = electronic Bohr magneton,

$$x = \frac{(g_j - g_1)\,\mu_o B}{h\Delta\nu},$$

g_1 = negative ratio of magnetic moment of nucleus to angular momentum expressed in units of μ_o,

g_j = a ratio analogous to g_1 applying to the electronic structure.

The shift in level, expressed in frequency units per unit change in B, may be found by calculating the derivative of $E_{i \pm \frac{1}{2}, m}$ with respect to B and dividing by h, yielding, for zero fields,

[25] G. Breit and I. I. Rabi, Phys. Rev. 38, 2082 (1931).

$$\nu'_{i \pm \frac{1}{2}, m} = L_o m \left[g_i \pm \frac{g_j - g_i}{2I + 1} \right] , \qquad (34)$$

where $L_o = \mu_o/h$ = the Lorentz unit, 1.400 Mc/gauss.

Since g_i is generally small compared to g_j (which is equal to 2), the spacings between adjacent levels in the upper set ($F = I + 1/2$) and lower set ($F = I - 1/2$) are essentially equal and have the value of $L_o/(I + 1/2)$ which for Cs^{133} is about 0.35 Mc/gauss.

The change in frequency in a line per unit change in B, neglecting second order terms, is found by subtracting the value of E' for the lower level from the value for the upper level. Numerical values for the components of the inner line pairs (which are doublets) and the second pair are shown in Table II. These are based upon the following values: $I = 7/2$, $g_j = 2$, $g_i = -2.558/1830 = -1.4 \times 10^{-3}$.

In the third column in Table II are shown the experimental values determined from the reciprocal slopes of the lines shown in Figure 11, which were calculated by the method of least squares. The first order theory predicts that the values of the two members of a symmetrical pair should be equal in magnitude. The fact that, in both cases, the higher frequency member of a pair had a value greater by about 5 per cent makes evident that second order terms in x cannot be neglected even for these small fields.

Accordingly the following analysis was made. A least square fit to data involving quadratic terms would be difficult and perhaps meaningless when the quadratic terms are small. However the only unknowns are the rate of variation of magnetic field with current in the Helmholtz coils, and the zero of the magnetic field. The former of these can be determined by the slope of the linear term and the latter by the intersection of the linear terms of the two pairs. Since the error in the value of the slope of the linear term, as computed from the dimensions of the coil, is not over a few per cent and since the quadratic terms are very small themselves, the quadratic terms were computed and subtracted from the experimental data. A least square fit was then made of the difference of the experimental data and the computed quadratic terms, and the slopes thus determined are shown in the fourth column of Table II. As can be seen, more than one half of the discrepancy has been removed by taking account of quadratic terms. The remaining discrepancy is probably not significant and may be due to nonlinear effects in the material of the cavity.

The theoretical values in Table II indicate that the doublet separation is about 0.1 per cent of the total deviation of the line; and calculations on other doublets not shown in the table give the same result. With the flux densities available, this effect should have been on the border of observation; an indication of the broadening of the doublet line is just noticeable. Undoubtedly, the difficulty in observing this effect is due to broadening of the lines because of inhomogeneities in the magnetic field.

TABLE II

Change in Frequency per Unit Change in Flux Density

Line	Change in frequency (Mc/sec) per gauss			
	Computed	Measured Linear Approximation	Measured Quadratic Approximation	Intercept with Quadratic Approximation
F = 4, m = −1 → F = 3, m = 0	−0.3524	-0.35_6	-0.36_3	9192.461
F = 4, m = 0 → F = 3, m = −1	−0.3483			
F = 4, m = 1 → F = 3, m = 0	0.3524	0.37_8	0.37_2	9192.558
F = 4, m = 0 → F = 3, m = 1	0.3483			
(−1 → −1)	−0.7007	-0.71_0	-0.71_8	9192.322
(1 → 1)	0.7007	0.74_7	0.74_2	9192.725

$\Delta\nu$ was determined from the intercepts as shown in the fifth column of Table II. The intercept frequencies of the outer pair, (1 → 1) and (-1 → -1), lines were determined with the working magnetic field vertical

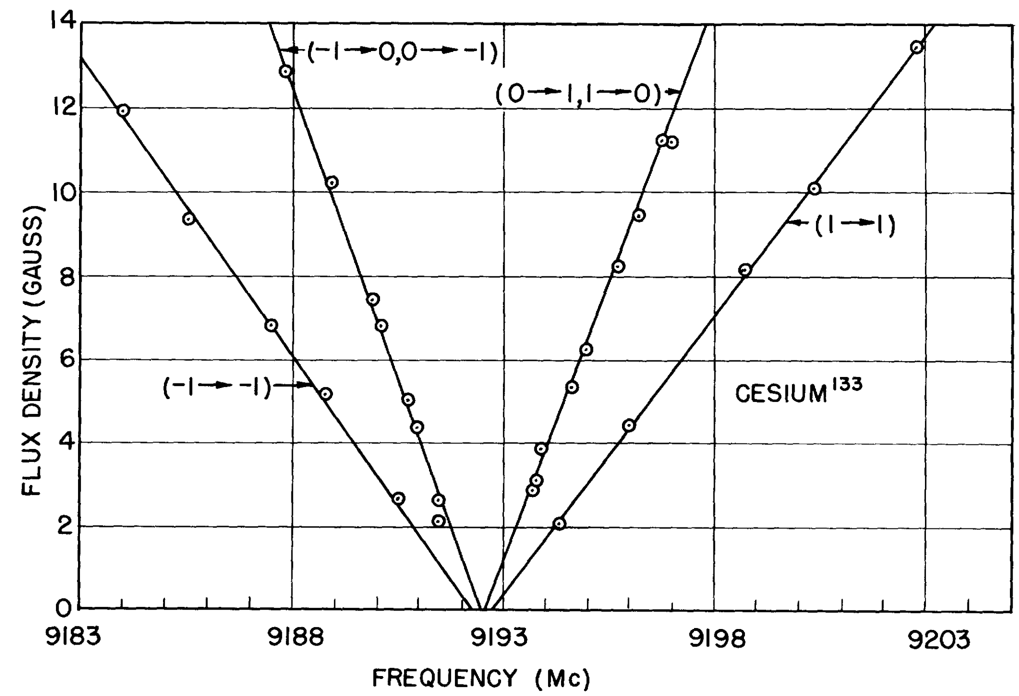

Fig. 11 Frequency dependence on field.

in order to favor σ components, and in each case the line shape was determined by averaging the values of flux density at the positive maximum and at the two zeroes, while the frequency was the average of values determined before and after the rough determination of line shape. It was discovered that the largest source of experimental error was frequency drift due to temperature changes of the cavity during the comparatively long time necessary to make even a crude determination of line shape. Therefore, when the inner pair were observed later (with the working field horizontal), the average of four or five values of flux density necessary to produce maximum deflection on the oscilloscope connected to the output of the tuned 30-cps amplifier was compared to the average of frequency measurements taken before and after. As the time required was much shorter, this method was more precise. Therefore, partly for this reason and partly because of the fact that the frequencies of these lines inherently vary the least with flux density, the extrapolated values for these inner lines are to be considered the more accurate.

Ideally, of course, these four intercepts should coincide. Their failure to coincide is to be attributed to stray fields or imperfect earth's field corrections. Such effects would affect the values from the two members of a symmetrical pair in equal and opposite amounts and could be eliminated by averaging the two values. The average values of the intercepts are 9192.510 Mc/sec and 9192.524 Mc/sec for the inner and outer pairs, respectively.

The rms deviation of the data from the least-square solutions of the inner lines was about 0.06_6 Mc/sec, and thus the probable experimental error of their intercepts is to be taken as 0.04_4 Mc/sec. For the reasons stated previously, the value due to the outer pair is to be considered as having an experimental error twice as large. A weighted average yields a value of $\Delta\nu = 9192.513$ Mc/sec with a probable error no greater than 0.030 Mc/sec. Since the inherent error in the frequency standard, as stated previously, is 0.006 Mc/sec, further accuracy could be achieved by obtaining more data or devising a better frequency-comparison process.

This value of $\Delta\nu$ is to be compared with 9192.6 ± 0.5 Mc/sec, the best previous value[6] obtained by atomic beam values, and is in excellent agreement. Since the atomic beam value was obtained from calculations based upon data at high magnetic fields and at low frequencies (about 2 Mc/sec), this agreement indicates that the accuracy of such calculations and data is remarkably good.

XII. CONCLUSIONS

This report has described a cavity method for observation of microwave spectra. It has been tested by study of the microwave atomic spectrum of Cs^{133}, and by measurements of the $\Delta\nu$ a check has been made on the work done previously by the atomic beam method. The present method requires a monatomic vapor not having a ground state $J = 0$, which practically limits its applicability in atomic spectra to the halogens and alkalies. However, in principle it may be used for the study of spectra of diatomic and polyatomic molecules. The use of this method can be extended by the use of a Stark effect modulation in place of Zeeman effect modulation by insulating one portion of the cavity from another without destroying its Q, so that the required external electric field can be applied. Its main advantage is the high sensitivity and ability to work with small samples. In the present experiment, a sample of 3×10^{15} atoms having an absorption coefficient of 3.5×10^{-8}/cm was employed; smaller samples or absorption coefficients could be used with a reduction in signal-to-noise ratio and with improvements in techniques which have been suggested. An important disadvantage, in addition to the limitation mentioned previously, is that observations require a long time and, therefore, in its present form this method is not suited for lines whose frequencies are not already approximately known.

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